

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:
Gary T. Rochelle and John Cullinane

Serial No.: 10/551,834

Filed: October 17, 2006

For: POLYAMINE/ALKALI SALT BLENDS
FOR CARBON DIOXIDE REMOVAL
FROM GAS STREAMS

Group Art Unit: 1797

Examiner: Ives J. Wu


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July 18, 2011
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Tamsen L. Barrett

APPEAL BRIEF

MAIL STOP APPEAL BRIEF - PATENTS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313

Dear Sir:

Appellants submit this Appeal Brief to the Board of Patent Appeals and Interferences in response to the final Office Action dated December 22, 2010. Appellants filed a Notice of Appeal on March 18, 2011. Accordingly, the deadline for filing the Appeal Brief is July 18, 2011, in light of the two month request for an extension of time submitted herewith. The required fees in connection with the filing of this Appeal Brief are being charged to a credit card through EFS-Web concurrently with this submission. Should any additional fees be required under 37 C.F.R. §§ 1.16 to 1.21, please consider this paragraph such a request and authorization

to withdraw the appropriate fee from Fulbright & Jaworski L.L.P. Account No.: 50-1212/UTSB:719US.

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I. REAL PARTY IN INTEREST

The real party in interest for this appeal is the assignee, The Board of Regents of the University of Texas System.

II. RELATED APPEALS AND INTERFERENCES

There are no related appeals or interferences.

III. STATUS OF THE CLAIMS

Claims 1-10, 17-26, and 28-42 are pending in this case and are rejected. Claims 11-16 and 27 have been canceled. The rejection of claims 1-10, 17-26, and 28-42 is being appealed.

IV. STATUS OF AMENDMENTS

No amendments are pending.

V. SUMMARY OF CLAIMED SUBJECT MATTER¹

Independent claim 1 is directed to a method of removing CO₂ from a gaseous stream comprising: contacting a gaseous stream with a solution (p. 3, lns. 4-5), the solution being formed by combining at least: a primary or secondary polyamine having an amine concentration of at least 4.0 equivalents/Kg water (p. 3, lns. 27-28), wherein the amines located on the polyamine are not sterically hindered (p. 4, lns. 21-24), an alkali salt having a concentration of at least 1.0 equivalents/Kg water (p. 3, lns. 6-7), and water (p. 3, ln. 7); wherein no monohydric or polyhydric alcohol is added to the solution (p. 4, lns. 7-8); whereby contacting removes CO₂ from the gaseous stream (p. 3, ln. 7); and regenerating the solution (p. 3, lns. 7-8).

Independent claim 17 is directed to a method of removing CO₂ from a gaseous stream comprising: contacting a gaseous stream with a solution (p. 3, lns. 22-23), the solution being

¹ Parentheticals citing to support in the specification for the claim language are exemplary and not meant to indicate that the specific citations are the only support in the specification for the claim language.

formed by combining at least: a primary or secondary polyamine having an amine concentration of at least 5.1 equivalents/Kg water (p. 3, lns. 24-25), wherein the amines located on the polyamine are not sterically hindered (p. 4, lns. 21-24), an alkali salt having a concentration of at least 5.1 equivalents/Kg water (p. 3, lns. 25-26), and water (p. 3, ln. 26); wherein no monohydric or polyhydric alcohol is added to the solution (p. 4, lns. 7-8); whereby contacting removes CO₂ from the gaseous stream (p. 3, lns. 26-27); and regenerating the solution (p. 3, lns. 26-27).

Independent claim 26 is directed to a method of removing CO₂ from a gaseous stream comprising: contacting a gaseous stream with a solution (p. 3, lns. 4-5), the solution being formed by combining at least: a primary or secondary polyamine having an amine concentration of at least 4.0 equivalents/Kg water (p. 3, lns. 27-28), wherein the amines located on the polyamine are not sterically hindered (p. 4, lns. 21-24), an alkali salt having a concentration of at least 1.0 equivalents/Kg water (p. 3, lns. 6-7), and water (p. 3, ln. 7); wherein no monohydric or polyhydric alcohol is added to the solution (p. 4, lns. 7-8); whereby contacting removes CO₂ from the gaseous stream (p. 3, ln. 7); and regenerating the solution (p. 3, lns. 7-8).

Independent claim 36 is directed to a method of removing CO₂ from a gaseous stream comprising: contacting a gaseous stream with a solution (p. 4, lns. 9-10), the solution being formed by combining at least: a piperazine derivative having an amine concentration of 4.0-10.0 equivalents/Kg water (p. 4, lns. 11-12), wherein the amines located on the piperazine derivative are not sterically hindered (p. 4, lns. 21-24), an alkali salt having a concentration of 4.0-10.0 equivalents/Kg water (p. 4, lns. 11-12), and water (p. 4, ln. 13); wherein the concentration of the piperazine derivative and the concentration of the alkali salt are approximately equal (p. 4, lns. 13-14); wherein no monohydric or polyhydric alcohol is added to the solution (p. 4, lns. 7-8);

whereby contacting removes CO₂ from the gaseous stream (p. 4, ln. 14); and regenerating the solution (p. 4, lns. 15).

VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Claims 1-10, 17-26, and 28-42 were rejected as obvious over US Pat. No. 5,017,350 to Hakka, *et al.* (“Hakka”).

VII. ARGUMENT

The Examiner concedes that Hakka fails to disclose the alkali salt concentrations recited in independent claims 1, 17, 26, and 36, but nevertheless contends that it would have been obvious to adjust the concentration of the alkali salt to meet the claimed concentrations “and thereby produce optimal working results.” Action mailed December 22, 2010 at ¶¶ 11, 22, 33, and 44. Applicants disagree that this reference renders the claims obvious.

A. The concentration of the alkali salt was not recognized as a result-effective variable

The Examiner is relying on case law allegedly standing for the proposition that “[t]he discovery of an optimum value of a known result effective variable, without producing any new or unexpected results is within the ambit of a person of ordinary skill in the art.” Action at, *e.g.*, ¶ 11 (citing *In re Boesch*, 205 USPQ (CCPA 1980), and MPEP § 2144.05). However, as set forth in MPEP §2144.05, a particular parameter must *first* be recognized as a result-effective variable, *i.e.*, a variable that achieves a recognized result, before the determination of the optimum or workable ranges of said variable might be characterized as routine experimentation. *In re Antonie*, 559 F.2d 618, 195 USPQ 6 (CCPA 1977). As recognized in several decisions by the Board of Patent Appeals and Interferences, the burden is on the Examiner to establish that a particular parameter is recognized as a result-effective variable in order to invoke “optimization”

case law. *See, e.g., Ex parte Johnson*, Appeal No. 2010-003975, Application No. 09/950,985, Decision on Appeal mailed 7/22/2010 (reversing obviousness rejection based on finding that Examiner failed to produce evidence that prior art recognized certain parameter as a result-effective variable). Contrary to the assertions of the Examiner, that burden has not been met in this case, nor could it be.

In the Final Office Action mailed December 22, 2010, the Examiner states that “the Examiner believes there would be instances where [varying the alkali salt concentration in the solution based on the parameters of the treated gas stream] would be necessary,” and labels alkali salt concentration as a recognized results-effective variable. Office Action mailed December 22, 2010, page 2. Applicant respectfully disagrees. First, the **only** mention of alkali salts in Hakka comes in the following brief paragraph:

The absorbing medium may contain mixtures of amine sorbents. Other components, e.g., alkali salts, may be present as additives to, for instance, retard sulphite or bisulphite oxidation, maintain pH and serve as cosolvents.

Hakka at col. 8, lines 15-19. Nowhere in that paragraph, nor anywhere else in Hakka, is there a recognition that the specific results listed in Hakka (*i.e.*, retarding sulphite or bisulphite oxidation, maintaining pH, and serving as a cosolvent) are dependent upon the concentration of alkali salt in the absorbing medium. Therefore, alkali salt concentration is not recognized as a result-effective variable for **any** result in Hakka, much less the specific results disclosed in the present application (which are not disclosed in Hakka, as explained below). Optimization of a parameter not recognized as being result-effective is not *prima facie* obvious. *In re Antonie*, 559 F.2d at 620.

B. Optimizing the concentration of alkali salt for the purposes of Hakka would not result in the concentration currently claimed

Moreover, even if there was a recognition in Hakka that the alkali salt concentration was a result-effective variable for the specific results listed in Hakka, the Examiner has not come forward with any evidence, nor could he, that optimizing the concentration of alkali salt for the specific results listed in Hakka would necessarily result in Applicants' claimed concentrations. This is because, as discussed in the present specification, Applicants include alkali salts in their absorbing medium for results that are different from those disclosed in Hakka. Specifically, as explained at pages 2 and 8-9 of the specification, it had been thought previously that concentrations of piperazine (PZ) greater than 1.3 m cannot be used in a CO₂-rich environment due to the formation of piperazine carbamate that precipitates from the solution. *See, e.g.,* U.S. Pat. No. 4,336,233 at col. 3, lines 7-18 (stating that “the use of piperazine alone as the washing agent is of limited applicability . . . [because] larger amounts than 1.3 mole / l cannot be used because in the presence of CO₂ . . . the carbamate of piperazine precipitates . . .”). Furthermore, the claimed concentrations of polyamine had been thought infeasible due to amine volatility. Specification at 2. However, Applicants discovered the surprising and unexpected result that greater concentrations of polyamines are feasible in a CO₂-rich environment without precipitation of solids and/or evaporation of the amine when used with certain concentrations of alkali salts because the interaction of the alkali with the amine and the CO₂ avoids solid precipitation and reduces the volatility of the amine, especially with PZ. *See* Specification at, *e.g.,* page 9, lines 6-11 (discussing result of avoiding solid precipitation) and page 14, lines 30-32 (discussing result of reducing amine volatility).

Nowhere does Hakka acknowledge or recognize that the higher concentrations of amine it allegedly discloses (which far exceed 1.3 m) were not thought to be possible with piperazine in

a CO₂-rich environment due to the formation of piperazine carbamate that precipitates from the solution with concentrations of piperazine greater than 1.3 m. In fact, Hakka appears to teach that the absorbing medium need not be single phase (*see* col. 7, lines 62-64), which confirms that Hakka does not even view solid precipitation generally as a problem needing to be solved. Furthermore, nowhere does Hakka recognize that the higher concentrations of polyamine it allegedly discloses had been thought infeasible due to amine volatility. Thus, it certainly cannot be argued that one of ordinary skill in the art would have had a reason, based upon Hakka, to “optimize” the alkali salt concentration to achieve the particular results of avoiding solid precipitation and reducing amine volatility. Again, optimization of a parameter not recognized as being result-effective is not *prima facie* obvious. *In re Antonie*, 559 F.2d at 620.

For at least these reasons, this rejection is flawed, and reconsideration and reversal thereof is respectfully requested.

VIII. CONCLUSION

For at least the reasons presented above, the rejections of the appealed claims should be reversed.

Respectfully submitted,



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IX. CLAIMS APPENDIX

1. A method of removing CO₂ from a gaseous stream comprising:
contacting a gaseous stream with a solution, the solution being formed by combining at least:
a primary or secondary polyamine having an amine concentration of at least 4.0 equivalents/Kg water, wherein the amines located on the polyamine are not sterically hindered,
an alkali salt having a concentration of at least 1.0 equivalents/Kg water, and
water;
wherein no monohydric or polyhydric alcohol is added to the solution;
whereby contacting removes CO₂ from the gaseous stream; and
regenerating the solution.
2. The method of claim 1, wherein the polyamine is piperazine, aminoethylpiperazine, hydroxyethylpiperazine, ethylenediamine, or dimethyl ethylenediamine.
3. The method of claim 1, wherein the alkali salt is potassium carbonate, sodium carbonate, lithium carbonate, a bicarbonate salt, a bisulfide salt or a hydroxide salt.
4. The method of claim 1, wherein the gaseous stream is contacted with the solution at a temperature of approximately 25°C.-120°C.
5. The method of claim 1, wherein the rate constant for the reaction of CO₂ with the primary or secondary polyamine (K_{PZ}) is at least 25 m³/mol-s at 25°C.
6. The method of claim 1, wherein the solution further comprises an additive.
7. The method of claim 1, wherein the polyamine concentration and the alkali salt concentration are at least 2.3 m.
8. The method of claim 1, wherein the ratio of equivalents of alkali salt to equivalents of polyamine is 0.3-3.0.

9. The method of claim 1, further comprising applying a water wash system, wherein the water wash system collects the polyamine from treated gaseous stream.
10. The method of claim 1, wherein the rate for the solvent-mediated removal of CO₂ from the gaseous stream is at least 1.5 times the rate for CO₂ removal in a method using an aqueous solution of 5.0-M monoethanolamine.
17. A method of removing CO₂ from a gaseous stream comprising:
contacting a gaseous stream with a solution, the solution being formed by combining at least:
 - a primary or secondary polyamine having an amine concentration of at least 5.1 equivalents/Kg water, wherein the amines located on the polyamine are not sterically hindered,
 - an alkali salt having a concentration of at least 5.1 equivalents/Kg water, andwater;
wherein no monohydric or polyhydric alcohol is added to the solution;
whereby contacting removes CO₂ from the gaseous stream; and
regenerating the solution.
18. The method of claim 17, wherein the concentration of the polyamine and the concentration of the alkali salt are at least 5.5 equivalents/Kg water.
19. The method of claim 17, wherein the concentration of the polyamine and the concentration of the alkali salt are approximately equal.
20. The method of claim 17, wherein the polyamine is piperazine, aminoethylpiperazine, hydroxyethylpiperazine, ethylenediamine, or dimethyl ethylenediamine.
21. The method of claim 17, wherein the alkali salt is potassium carbonate, sodium carbonate, lithium carbonate, a bicarbonate salt, a bisulfide salt or a hydroxide salt.
22. The method of claim 17, wherein the gaseous stream is contacted with the solution at a temperature of approximately 25°C-120°C.

23. The method of claim 17, wherein the rate constant for the reaction of CO₂ with the primary or secondary polyamine (K_{PZ}) is at least 25 m³/mol-s at 25°C.
24. The method of claim 17, wherein the solution further comprises an additive.
25. The method of claim 17, wherein the rate for the solvent-mediated removal of CO₂ from the gaseous stream is at least 1.5 times the rate for CO₂ removal in a method using an aqueous solution of 5.0-M monoethanolamine.
26. A method of removing CO₂ from a gaseous stream comprising:
contacting a gaseous stream with a solution, the solution being formed by combining at least:
 - a primary or secondary polyamine having an amine concentration of at least 4.0 equivalents/Kg water, wherein the amines located on the polyamine are not sterically hindered,
 - an alkali salt having a concentration of at least 1.0 equivalents/Kg water, and
 - water;wherein no monohydric or polyhydric alcohol is added to the solution;
whereby contacting removes CO₂ from the gaseous stream; and
regenerating the solution.
28. The method of claim 26, wherein the polyamine is piperazine, aminoethylpiperazine, hydroxyethylpiperazine, ethylenediamine, or dimethyl ethylenediamine.
29. The method of claim 26, wherein the alkali salt is potassium carbonate, sodium carbonate, lithium carbonate, bicarbonate salt, a bisulfide salt, or a hydroxide salt.
30. The method of claim 26, wherein the gaseous stream is contacted with the solution at a temperature of approximately 25°C-120°C.
31. The method of claim 26, wherein the rate constant for the reaction of CO₂ with the primary or secondary polyamine (K_{PZ}) is at least 25 m³/mol-s at 25°C.
32. The method of claim 26, wherein the solution further comprises an additive.

33. The method of claim 26, wherein the polyamine concentration and the alkali salt concentration are at least 2.3 m.
34. The method of claim 26, wherein the ratio of equivalents of alkali salt to equivalents of polyamine is 0.3-3.0.
35. The method of claim 26, wherein the rate for the solvent-mediated removal of CO₂ from the gaseous stream is at least 1.5 times the rate for CO₂ removal in a method using an aqueous solution of 5.0-M monoethanolamine.
36. A method of removing CO₂ from a gaseous stream comprising:
contacting a gaseous stream with a solution, the solution being formed by combining at least:
a piperazine derivative having an amine concentration of 4.0-10.0 equivalents/Kg water, wherein the amines located on the piperazine derivative are not sterically hindered,
an alkali salt having a concentration of 4.0-10.0 equivalents/Kg water, and
water;
wherein the concentration of the piperazine derivative and the concentration of the alkali salt are approximately equal;
wherein no monohydric or polyhydric alcohol is added to the solution;
whereby contacting removes CO₂ from the gaseous stream; and
regenerating the solution.
37. The method of claim 36, wherein the piperazine derivative is piperazine, aminoethylpiperazine, or hydroxyethylpiperazine.
38. The method of claim 36, wherein the alkali salt is potassium carbonate, sodium carbonate, lithium carbonate, a bicarbonate salt, a bisulfide salt, or a hydroxide salt.
39. The method of claim 36, wherein the gaseous stream is contacted with the solution at a temperature of approximately 25°C-120°C.

40. The method of claim 36, wherein the rate constant for the reaction of CO₂ with the piperazine derivative (K_{PZ}) is at least 25 m³/mol-s at 25°C.
41. The method of claim 36, wherein the solution further comprises an additive.
42. The method of claim 36, wherein the rate for the solvent-mediated removal of CO₂ from the gaseous stream is at least 1.5 times the rate for CO₂ removal in a method using an aqueous solution of 5.0-M monoethanolamine.

X. EVIDENCE APPENDIX

None.

XI. RELATED PROCEEDINGS APPENDIX

None.